LA-UR-20-30314

Title: FY2020 Progress Report

Author(s): Bredeweg, Todd Allen

Intended for: Report

Issued: 2020-12-17

FY2020 Progress Report

Project: Integral Measurements of Independent and Cumulative Fission Product Yields Supporting Nuclear Forensics and Other Applications

Project Number: LA19-ML-Integral_Fission_Product_Yield-NDD3Ad

Principal Investigator: Todd Bredeweg, LANL, (505) 667-2246

HQ Project Manager: Timothy Ashenfelter

Date submitted: 14 December 2020

Summary (LANL/LLNL/PNNL)

Activities for most tasks continued smoothly from FY19 into FY20. A Science Plan was developed for the F2019 Nuclear Forensics Venture in Q1, and was presented at HQ on Jan17, 2020. The Science plan included input from all tasks in the F2019 Venture plus this project, and was assembled and presented jointly by all three labs.

Several experimental and theoretical fission product yield efforts were funded out of the 2018 Interagency FOA, and all of them have built-in interconnections to ensure that data collected by the experimental projects are handed off to the theory and evaluation for incorporation into the international nuclear data libraries and future fission yield evaluations. A requirement in one of the LANL theory projects was to coordinate a workshop to facilitate these efforts. The International Workshop on Fission Product Yields (FPY2019) was held at the Inn and Spa at Loretto in Santa Fe, NM 9/30 – 10/3, 2019. This was followed by a one day closed session that was held at LANL on 10/4/2019. Proceedings for the Santa Fe sessions will be published. Todd Bredeweg was on the local organizing committee, and Bruce Pierson was a member of the international organizing committee.

Work performed under this project was presented at Nuclear Data Week 2019, historically sponsored and hosted by the National Nuclear Data Center at Brookhaven National Laboratory. There was some lively discussion, and we were invited to participate in an IAEA Collaborative Research Project (CRP) focused on the upcoming re-evaluation of fission product yields for the next major release of the ENDF evaluated nuclear data library.

The SLFPY run on the Godiva critical assembly that focused on ^{237}Np was executed, after several delays, the week of Jan 20, 2020. Data return was good, and a preliminary report was submitted to HQ. Data analysis is nearing completion, and a final report and publication are imminent.

The Workshop for Applied Nuclear Data Activities (WANDA 2020) was held at the Elliott School of International Affairs at George Washington University March 3-5, 2020, followed by a one-day invitation-only classified session on January 6, 2020. This workshop was modeled after the Nuclear Data Road-mapping and Enhancement Workshop (NDREW), held at the UCDC center in 2018, and WANDA 2019, held at the Elliott School of International Affairs at George Washington University from January 22-24, 2019. Both helped to guide the Nuclear Data Interagency Working Group Funding Opportunity

Announcements that were released several months after the respective workshop. The purpose of the workshops is to discuss nuclear data needs and potential solutions for nuclear energy, nonproliferation, isotope production, and stewardship science among other important mission areas that are supported by U.S. Federal Agencies. The current project to study integral fission product yields was one of several fission product related efforts funded as part of the 2018 Interagency FOA, and it seemed appropriate to continue our participation in the efforts to coordinate investments in nuclear data research. Attendees from the Nuclear Data teams included Todd Bredeweg from LANL; Jason Burke from LLNL; and Larry Greenwood, Lori Metz and Bruce Pierson from PNNL, along with several others. Reports and proceedings for each of these workshops can be found at https://www.nndc.bnl.gov/ndwg/.

Starting near the end of FY20Q2 the local, state, national and international efforts to mitigate the spread of COVID-19 lead to forced work-from-home, resulting in considerable disruption to our programmatic and deliverable schedules. All experimental work on this and most other projects was put on hold from mid-March through the beginning of June. Attempts were made to continue progress during the extended period of "work from home", but this is not always efficient for an experimental project, where much of the work requires significant time in the laboratory. However, by mid-June some limited on-site work was starting at LANL and PNNL, and at LLNL several weeks later, and more concrete progress was being made on most tasks. There still are, and will continue to be challenges due to the extended, multi-lab (and multi-state) nature of much of the work, as we attempt to coordinate activities between institutions that are not necessarily on the same schedule, or under the same state rules. These difficulties will likely continue to slow progress until a vaccine is generally available. Overall, I had estimated that deliverables associated with experimental campaigns under this project would be postponed 6-9 months. This is true for our planned experiments at NCERC where scheduling and coordination can be more challenging. However, in a truly impressive feat of planning and execution, as wild fires raged across the Pacific North West, the second joint irradiation at the PNNL D-T (14 MeV) neutron source was completed in September, with aliquots of the dissolved HEU and DU samples being analyzed by both PNNL and LANL.

Task 1: Cumulative Fission Product Yields (LANL/PNNL)

Analysis of the samples from the August 2019 14 MeV irradiation of ^{238}U on the PNNL D-T neutron source was completed and a report of the results was submitted to HQ in December 2019. The report included final results for the PNNL and LANL FY19 R-value measurements for the 14 MeV neutron irradiation of DU and HEU along with a comparison to previous PNNL campaigns, AWE measurements, and literature values. Recommendations were made based on these results for future campaigns, and planning for the coordinated irradiations of ^{235}U on Flattop and the PNNL D-T neutron source in FY20 was initiated based on these recommendations. It was recognized that the logistics for this measurement campaign would be challenging, and would test both laboratories. Largely due to the additional complications, we had initially settled on scheduling the irradiations in late August or early September. Unfortunately, the COVID-19 mitigation efforts initiated in mid-March forced the team to postpone the Flattop irradiation to FY21. This decision was driven in part by the travel requirements to complete the fission chamber testing in advance of the irradiation. Since the DT portion of the measurement did not require a fission chamber it was decided to not postpone. This task was able to restart limited operations in Q3, which allowed staff to begin preparations for the irradiation campaign.

During the FY19 campaign, a reduced total neutron output from the D-T generator head was observed. A new D-T head was ordered in early Q3 and installed in Q4 before the scheduled irradiation.

Project: Integral Measurements of Independent and Cumulative Fission Product Yields Supporting Nuclear Forensics and Other Applications

On September 11/12, 2020 the cumulative fission product yield task conducted an irritation of HEU and DU with 14 MeV neutrons in the PNNL low scatter room using a DT neutron generator. The new DT head on the Thermo instrument was successfully installed and characterized just prior to this campaign. This year, the targets were 1.2g of HEU and 1.8g of DU (both 99% isotopically pure). The targets were prepared in coordination with LANL, and after irradiation, the solutions were split with LANL receiving ½ of the DU target and 1/3 of the HEU target, shown in Figure 1. (We were only able to ship non accountable quantities of HEU, thus the lower amount shipped to LANL).

Figure 1: Dissolved HEU target.

The splits for LANL were shipped on Monday September 14th after screening and dissolution over the weekend. The samples arrived at LANL on time and without any delays or issues with shipping.

Radiochemistry started at PNNL on Monday September 14th, however due to wildfires raging thought the west coast, the smoke blanketed PNNL and the laboratory was closed for 3 days, which was the Monday through Wednesday that the radiochemical separations were supposed to occur. We were able to work with laboratory management and get our radiochemistry staff labeled "essential" which allowed them to come into the radiochemistry building and proceed with the separations. It was challenging to organize this significant effort in the COVID-19 environment and adding the wildfire smoke restrictions on top of that at short notice required close coordination with our management and facility staff.

Radiochemical separations were completed by the end of Wednesday and all elements were on the counters. Samples will continue to count into FY21 year and the data will be compiled and shared between LANL and PNNL in preparation for the quarterly "meeting" on November 6th. An example of preliminary data is shown in Figure 2. In addition, one unexpected result from this campaign was the first direct measurement of Zn/Ga-72.

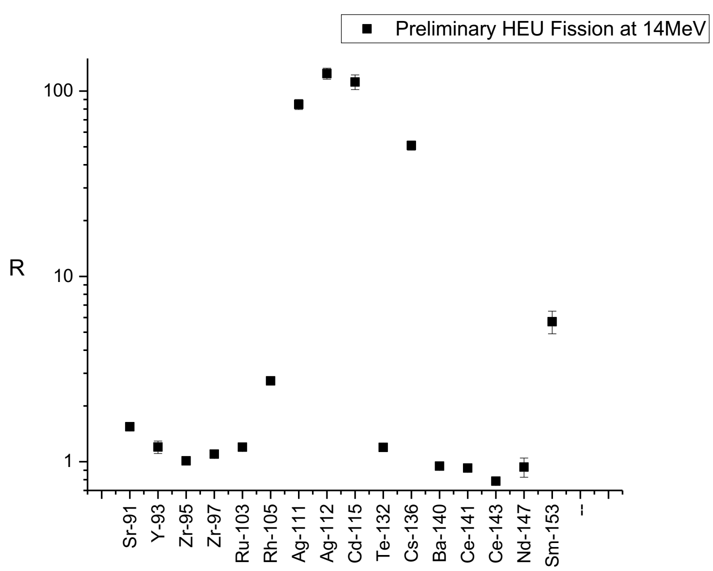

Figure 2: Preliminary R values measured by gamma spectroscopy from HEU.

Project: Integral Measurements of Independent and Cumulative Fission Product Yields Supporting Nuclear Forensics and Other Applications

Fission Chamber Development:

Final development and testing of the Flattop fission chambers was planned to follow a series of steps, each with a defined goal, including:

1. Benchtop testing using ^{252}Cf fission sources, the standard for testing the operation of fission detectors, to explore the effects of fill gas pressure (P-10 or CF_4) and bias voltage. Unfortunately, the yield of alpha particles (96.9% of decays) vs fission fragments (3.1% of decays) results in a large alpha background in the pulse-height spectrum. An ^{241}Am source can be used to get a measure of the detector response just to alpha particles.
2. Testing at a thermal neutron source (research reactor) using ^{235}U reference foils to determine the detector response just to fission products. The yield of alpha particles from spontaneous decay of ^{235}U is several orders of magnitude lower than ^{252}Cf, and thus we can obtain a much cleaner pulse-height spectrum for fission products.
3. Testing on Flattop using ^{235}U reference foils to determine the γ-ray background picked up in the fission chambers as a function of power level. The γ-rays arise from fission in the tens of kilograms of Oralloy fuel of the Flattop assembly that surrounds the fission chamber. This “stress test” is used to determine the power level above which the accidental summing begins to seriously affect the fission data.

Procurement, fabrication and authorization delays in FY19 resulted in doing some of the above out of order. Once we had everything in place to begin testing, a set of two modified fission chambers and the gas fill manifold to fill them, we went straight to the McClellan Nuclear Research Center (MNRC) outside Sacramento, CA.

Nine ^{235}U reference foils were shipped to the MNRC along with two modified fission chambers (FCs) and associated data acquisition hardware the week of September 9th for testing that was conducted the week of September 16th. The modified FCs were designed to enable recovering and swapping the ^{235}U reference foils during the tests rather than having to permanently dedicate a complete FC to each reference foil. While allowing for swapping reference foils the internal dimensions and layout of the modified FC are identical to those of the Mark II Flattop FCs, as shown in Figure 3. A photo of the modified FC is shown in Figure 4. A photo of the new gas fill manifold, used to pressurize the FCs, is provided in Figure 5. The gas fill manifold was also shipped to the MNRC.

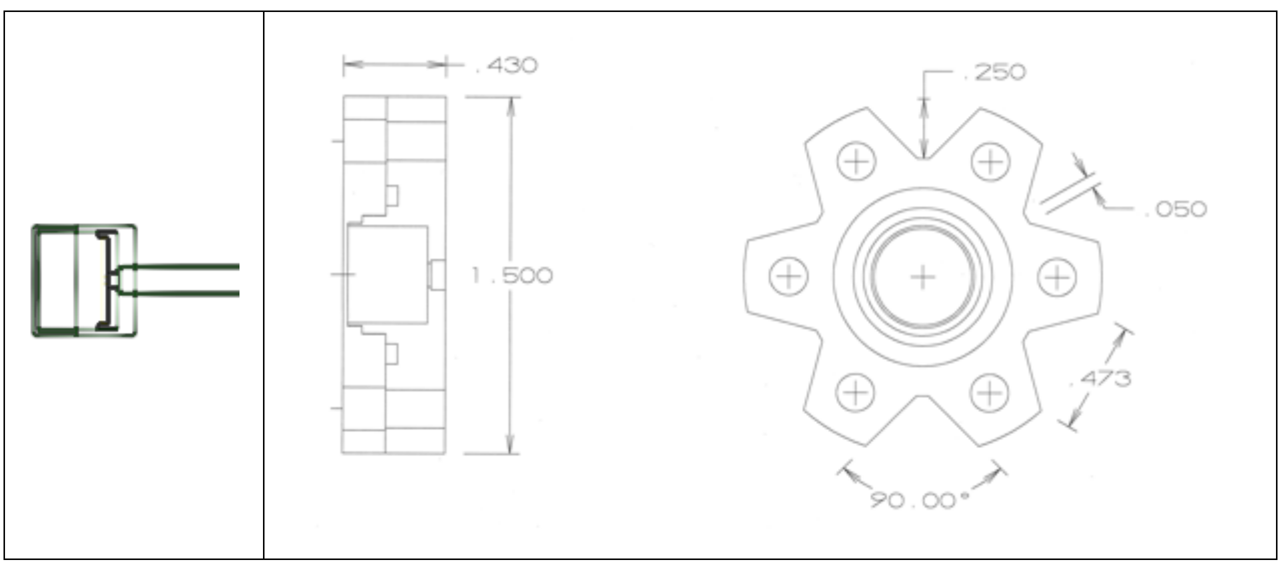

Figure 3: Drawings of the Mark II fission chamber "head" (left panel) that has been used on the Flattop critical assembly, and the "head" of the modified fission chamber (right panel) that will be used for testing reference foils, and later the anode-reference foil spacing.

Figure 4: Photo of the gas control body of the modified fission chamber (left) and the Mark II fission chamber (right).

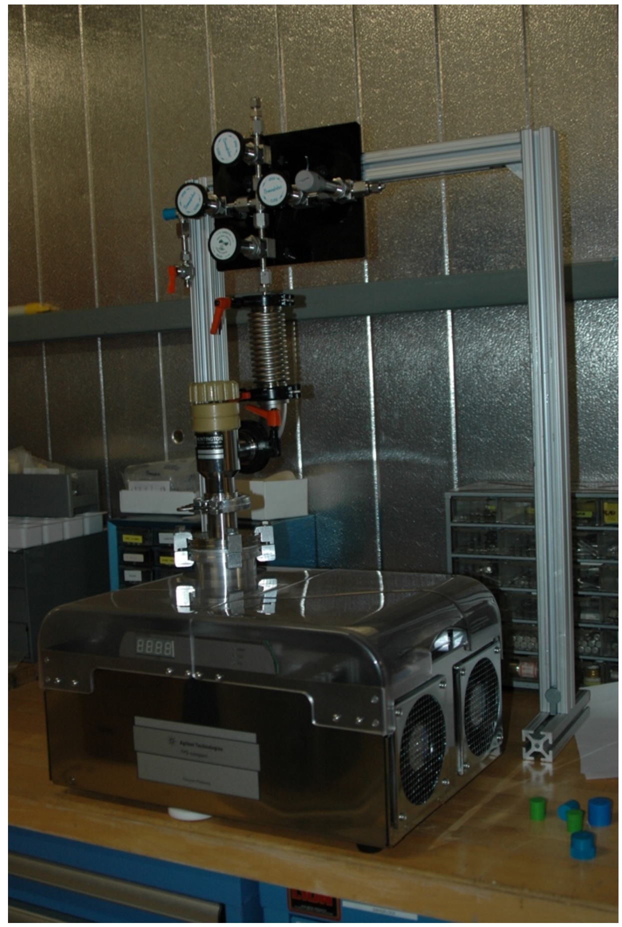

Figure 5: Photo of the new gas fill manifold.

As expected, there was a learning curve for doing work at a new facility, but the local staff were somewhat helpful and accommodating. We completed setup on the first day and were able to take data on two of the ^{235}U reference foils plus a blank over the next two days. Alas, in each case we observed the same distorted pulse height spectrum with the ^{235}U foils that were observed at NCERC in April 2018. Varying the P-10 pressure (80, 50 and 30 psig) and bias voltage (+100-400 V) had little effect.

After returning to LANL, and having several discussions on our observations, it became increasingly clear that the problem was most likely the spacing between the anode and the ^{235}U reference foil. We submitted drawings to our machine shop to fabricate a set of PEEK spacers to reduce the anode-reference foil gap to ½ and ¼ of the original. Benchtop testing started with the ½ gap PEEK spacers, providing an anode-reference foil gap of 0.34 cm, and a 2500 Bq ^{252}Cf source. The resulting pulse height spectra, shown in Figure 6, shows excellent alpha-fission product separation. The data in the plot were collected for bias voltages between 50 and 200 V with a P-10 gas pressure of 60 psi. The separation between the heavy and light mass peaks, and the alpha-fission product separation, improves dramatically with increasing bias voltage. Unfortunately, it was about this time, mid-March, when LANL moved to min-safe operations, halting all laboratory work. The lock-down was gradually eased starting in early June, and we were able to collect additional data using a P-10 gas pressure of 30 and 90 psi, and for bias voltages up to 450 V for both the ½ (0.34 cm) and ¼ (0.17 cm) gap spacers. The final benchtop tests will be a (partial) repeat of the above configurations using CF_4 gas in place of P-10. We ordered a cylinder of CF_4 in June, and finally received it in mid-November.

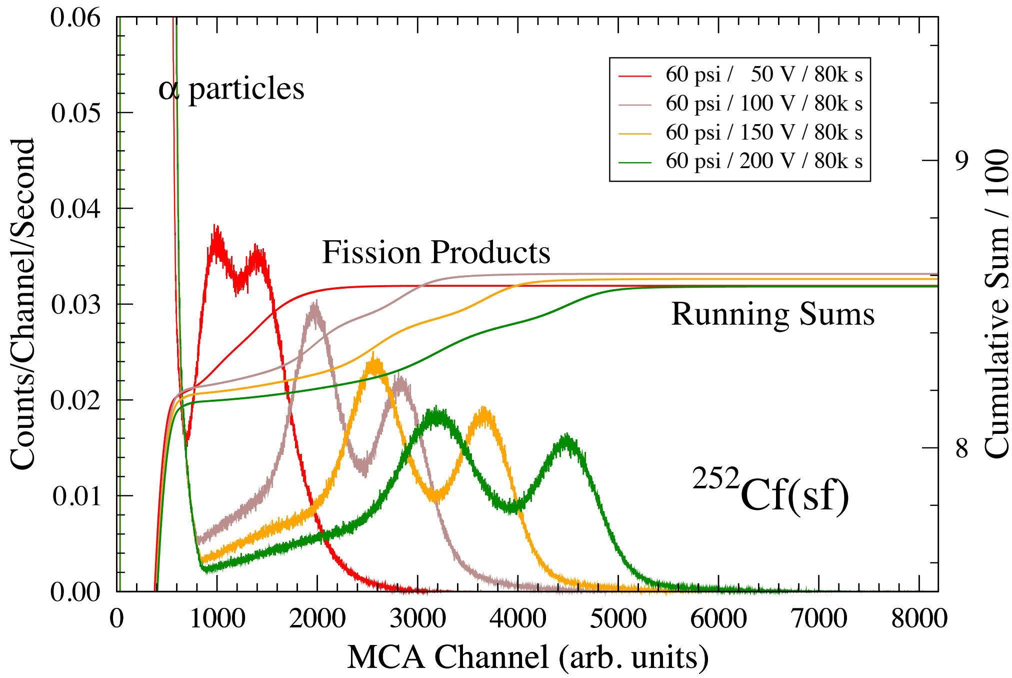

Figure 6: Fission chamber performance as a function of applied voltage using a 2500 Bq ^{252}Cf source and a gap of 0.34 cm. All data were taken using a P-10 gas pressure of 60 psi.

The final two phases of the FC testing will be conducted at the MIT Nuclear Research Reactor using ^{235}U reference foils (executed in late October) followed by similar tests on the Flattop assembly (scheduled for the week of Nov 16, but canceled due to shipping/receiving snafus that were the result of COVID-19 infections and mitigation efforts.).

By late October, 2019, several weeks after our return from the MNRC we had planned the following schedule for the remainder of FY20.

Benchtop testing with ^{252}Cf sources.	Anticipated completion by mid-April, 2020
FC testing with ^{235}U reference foils at the MIT Nuclear Reactor Center	Scheduled for mid-May, 2020
FC testing with ^{235}U on the Flattop assembly at NCERC	Scheduled for late June, 2020
Production run with ^{235}U on the Flattop assembly at NCERC	Scheduled for mid-September, 2020

Unfortunately, with the arrival of COVID-19 this all went out the window. We completed most of the benchtop testing over the summer, but did not receive the CF_4 until mid-November. Given the moving target of COVID mitigation requirements for New Mexico and Massachusetts we were barely able to complete the tests at the MIT Research Reactor, but were forced to cancel the tests on Flattop. At this time, we are trying to reschedule the tests on Flattop at least two weeks in advance of the production run that is currently scheduled for the week of April 12, 2021.

Sample Containment:

For NCERC sample irradiations DU, HEU and Np metal samples have all been wrapped in aluminum foil (0.01 mm, 99.35 % pure) and then sealed in aluminum capsules (Figure 7, left). This sample containment has worked very effectively, the tightness of the fit between lid and base forming an effective seal. However, the increased radiological hazards associated with plutonium necessitate more rigorous sample containment than a physical seal. Ultrasonic welding would provide a metallurgical bond between two pieces of aluminum foil and thus a more complete sample seal, as initially envisaged in Figure 7, right. This is the method of sample containment that is being actively pursued at LANL, with the goal of developing and testing this methodology with U targets prior to containment and irradiation of Pu targets.

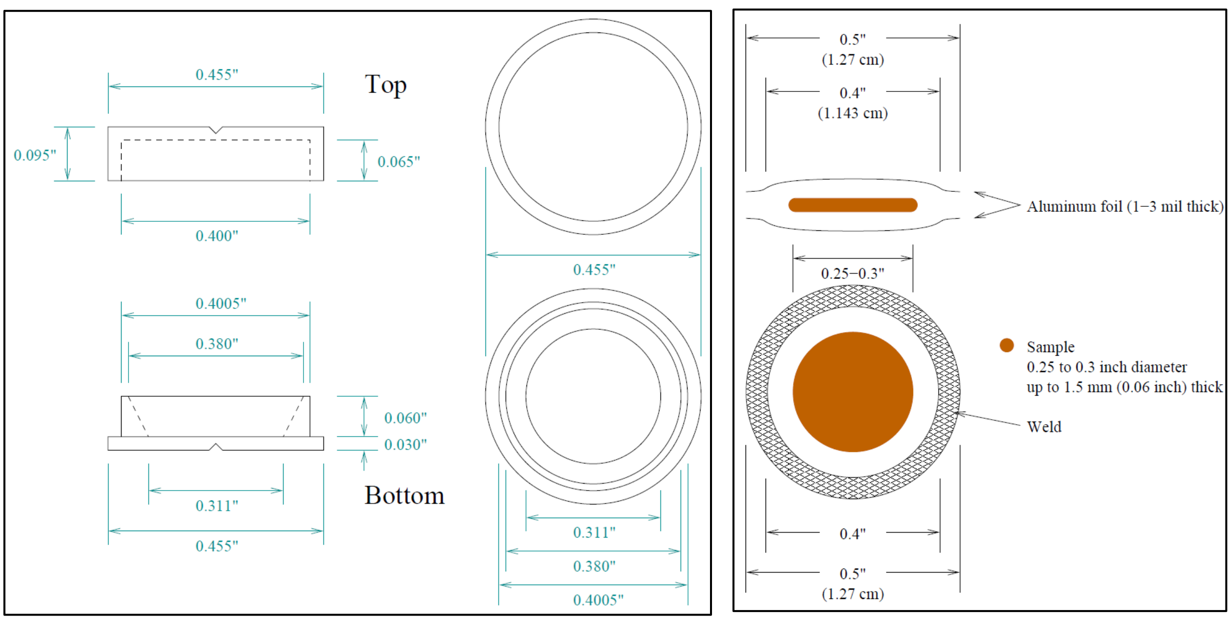

Figure 7: Actinide encapsulation, both current capsule method (left) and proposed welded method (right).

Previously, an Ultraweld L20 had been purchased (FY 2014) with the aim of containing actinide targets between shaped aluminum foil by ultrasonic welding (Figure 8). In FY 2019 it was determined that there were several components missing from the system, which also required configuration to our specific requirements. The Ultraweld L20 unit was thus returned to the manufacturer, Branson Ultrasonic Corporation (Danbury, CT) for servicing and re-tooling. Two LANL radiochemists visited the company at this time to assist in the development of the appropriate tooling and settings to meet expected requirements. The unit was received back at LANL at the end of the FY.

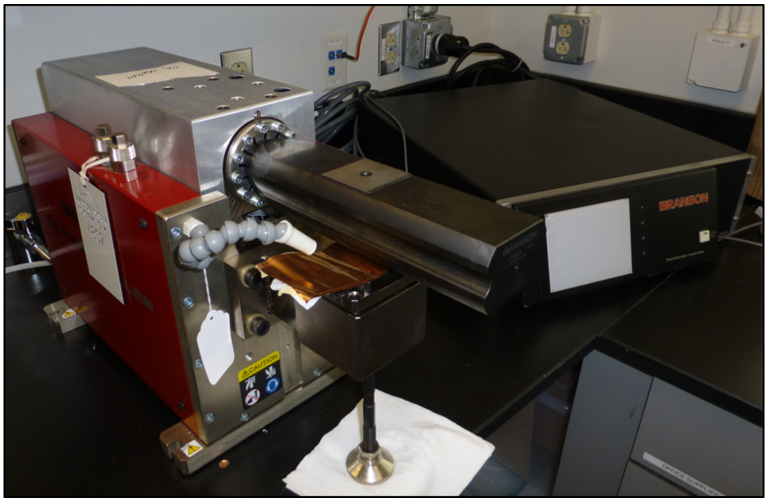

Figure 8: Branson Ultraweld L20.

Prior to testing the Ultraweld L20, the operation of this instrument was determined to be a new work activity, necessitating an internal safety review process prior to authorization being granted to operate the instrument. It should be noted that this authorization extends to the eventual sealing of plutonium samples. The system was set up in a non-rad area for initial development work, with the actuator connected to a compressed air cylinder and the power unit connected to the appropriate 30 Amp outlet. It would appear that a 20 Amp outlet is all that is required for this specific application, which would require the purchase of a new control unit. Switching out the control unit to change the electrical power requirements would greatly simplify the logistics associated with sealing plutonium samples.

Currently the Ultraweld L20 is set up to weld two pieces of aluminum into a disc with a pocket diameter of 0.455”, and will effectively weld two pieces of 0.003”, 0.004”, 0.005” and 0.006” aluminum shim. The lower thickness material (0.001”, 0.002” & 0.003”) is too thin, the effort of welding disintegrating the foil, and at >0.007” an effective weld is not possible. It should be noted that changing the energy, weld pressure or amplitude could increase the range of material thicknesses that can be effectively welded. Currently the horn component of the weld unit is completely flat with only the lower anvil piece possessing the raised ring weld surface (Figure 7, left) with internal indentation. This configuration provides distinct benefits, the sample can potentially be held completely within a lower pocket without the necessity of accurately aligning an upper pocket. However, this has created a challenge associated with generating a deep enough pocket in one piece of aluminum foil to contain target material. Several machine hand-tool configurations have been developed in an attempt to define a pocket size of at least 0.26” diameter and 0.07” depth. It has proven comparatively easy to rip the aluminum foil and thus challenging to prepare sample containment of a consistent standard. 1000 series aluminum alloy (> 99 % aluminum, with Cu, Mn, Si, Fe & Zn) is significantly more malleable, and can thus be more readily shaped. This may be the more appropriate material to use for containment if capsule integrity can be maintained. Current efforts are focused on preparing prototype capsules with copper discs sealed inside to simulate the actinide targets.

Project: Integral Measurements of Independent and Cumulative Fission Product Yields Supporting Nuclear Forensics and Other Applications

Target Removal Chemistry:

The presence of excess target material from an irradiated plutonium target, both Pu isotopes and ^{241}Am, can significantly impact the radiochemistry required to generate purified isotopes for CFPY (Cumulative Fission Product Yield) determination. In FY2020 the Radiochemistry Team determined, and validated, protocols that will allow for the analysis of purified CFP's in the absence of target activity interference.

In a future NCERC Nuclear Physics irradiation the plutonium target will be dissolved and the resultant "A" solution split *ca.* 50:50 between LANL and PNNL for duplicate analysis. Two 4 M HNO_3 Pu solutions prepared from a previously irradiated Pu target was used to validate the developed analysis plan, constructed from a combination of tried and tested LANL radiochemical procedures and additional Pu/Am removal processing steps. The solutions contained 4 mg and 44 mg of Pu, respectively which would correspond to a hypothetical initial *ca.* 100 mg target.

Irradiated NCERC samples typically return to LANL within a few days of irradiation which allows for the analysis of a suite of CFP's down to half-lives of < 1 day by separated radiochemical analysis. Due to the variation in CFPY across the peak and wing/valley mass no's a split into small (peak) and large (wing/valley) sample aliquots allows for the most efficient sample analysis, and in this case corresponding to the 4 and 44 mg Pu content solutions, respectively. The peak "Mo-side" sample contained Sr, Ba, Mo, Zr, Nd, Ce & Y carriers while the wing/valley "Ag-side" sample contained Ag, Cd, Cs & Sm, Eu & Tb carriers. The initial process step involved carrier equilibration and cation exchange column removal of the excess Pu target from both solutions, but not the removal of ^{241}Am. Subsequent sequential chemistries and separated element radiochemistry produced samples for counting, with additional clean up steps sometimes required to remove remaining transuranic activity. In the case of the rare earth elements (REE, Nd/Ce/Y and Sm/Eu/Tb) an additional ^{241}Am removal column was required prior to separated element chemistry.

In all cases the only sample activity that could be measured in the purified element mounts related to long-lived CFP from the prior irradiation (^{90}Sr, ^{137}Cs, ^{155}Eu & ^{144}Ce). The results indicate that it could be possible to undertake the complete standard suite of CFP analysis using the combination of beta counting/gamma spectrophotometry typically employed, and this extends to ^{147}Nd which behaves chemically very similarly to ^{241}Am. Gravimetric yields ranged between 20-55%, all acceptable for data collection. The only major concern is the significant time commitment required to undertake the Pu removal steps which could impact the analysis quality of the shortest half-life CFP's, ^{97}Zr and ^{143}Ce specifically. A diagram of the overall radiochemical process is shown in Figure 9.

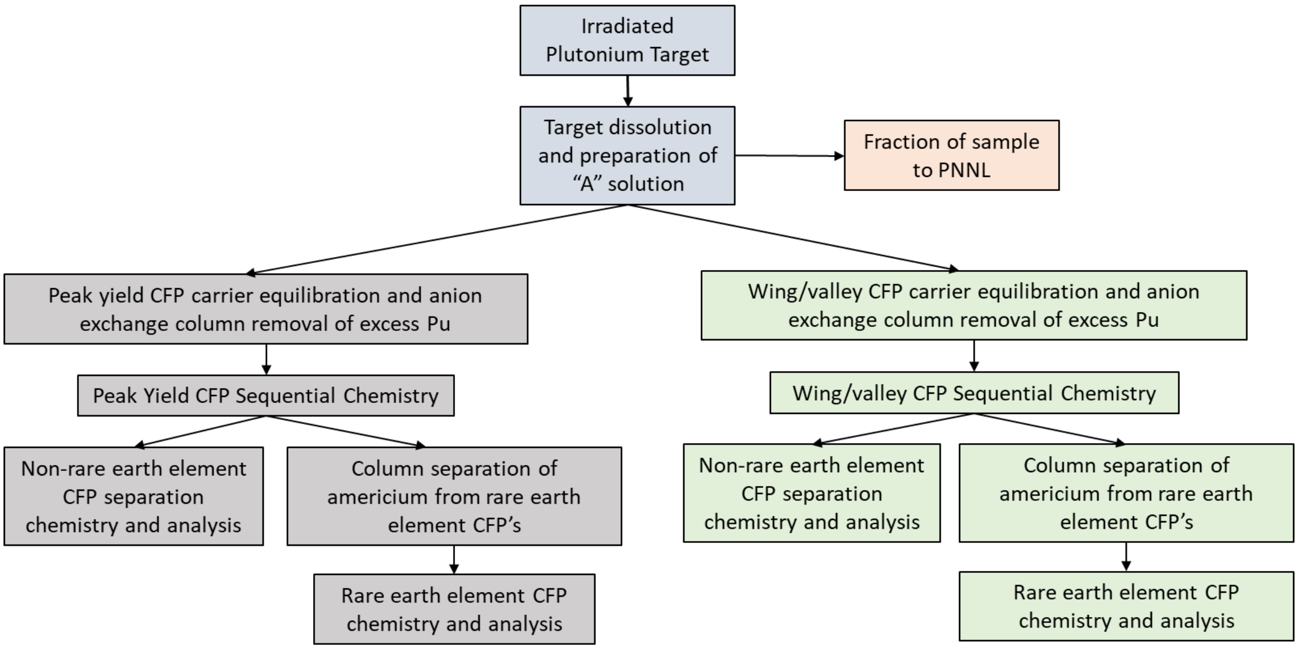

Figure 9: Radiochemical Separation Process from a Pu target.

Task 2: Cross Sections and Reaction Network Physics (LANL)

Personnel changes resulted in a temporary lull in activity, but progress resumed right before the LANL directive to work at home. We received the vacuum chamber for the new silicon drift detector (SDD) in September, and we have started to assemble the SDD counter. New alpha and gamma calibration sources were received in early FY20, and we have assembled a rigorous calibration plan for the new SDD and completed calibrations of several other radiation counters that we will use for this task and Task #1.

Task 3: Short-lived Fission Product Yields (LLNL/PNNL)

The Short-lived Fission Product Yields measurement of ^{237}Np on the Godiva IV critical assembly at NCERC was executed the week of January 20, 2020. Gamma ray spectroscopy of the irradiated samples continued for one week.

Staff at LLNL and PNNL continue to refine and improve on the measurement results from the ^{239}Pu and most recent ^{237}Np experiments. For the latest experiment, LLNL measured the detection efficiency using a suite of new single-isotope standards and PNNL using a NIST traceable mixed gamma standard. The results of the collaborative efficiency evaluation are provided below in Figure 10. Overall agreement is promising with some slight differences that are currently under review. Evaluation of the self-attenuation correction has also been independently verified by both laboratories. Staff at LLNL have identified that the use of the ^{233}Pa gamma-lines may cause issues in cases where the daughter is not in equilibrium with the ^{237}Np. Measured fission yields from the long-lived target show good agreement with ENDF-VIII.0. Fission yield measurements from the ^{237}Np irradiation are ongoing with LLNL and PNNL staff independently working up final results for comparison later this year.

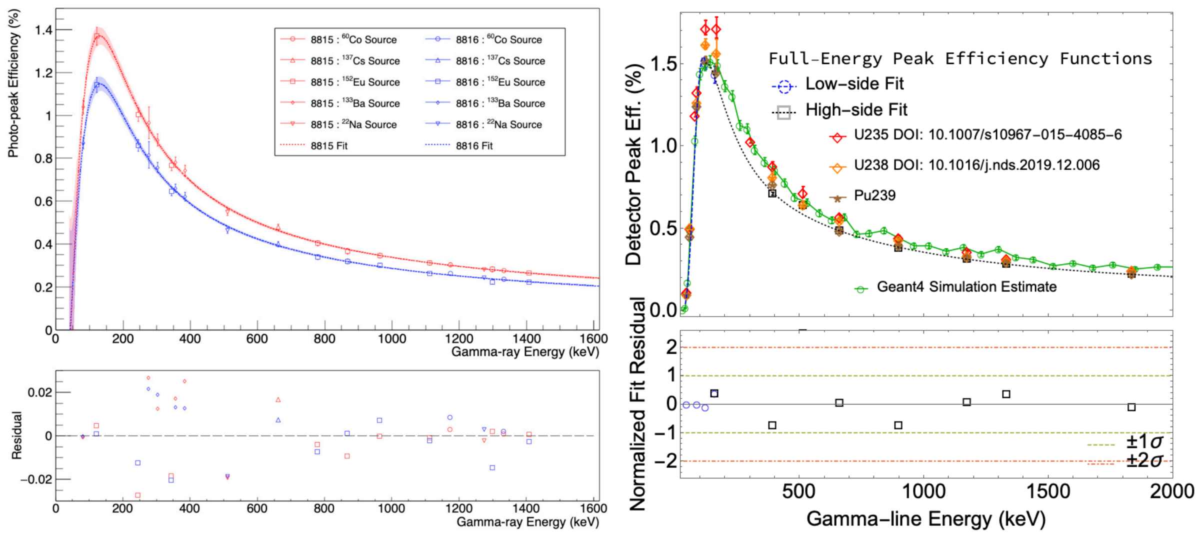

Figure 10: LLNL (left) and PNNL (right) measured detection efficiencies for the two detectors.

Preliminary measurements of the fission product yields from the ^{237}Np irradiation indicate improvements to the known fission yields and their uncertainties can be obtained for ^{93}Y, ^{104}Tc, ^{107}Rh, ^{129}Sb, ^{131}Sb, ^{133}I, and ^{138}Xe.

In addition to finalizing the detection efficiencies and associated gamma spectroscopy corrections, the neutron fluence was evaluated using a witness foil package composed of Fe, Co, Ni, Al, Cu, Au, and Ti. The addition of Al and Ni to the neutron fluence witness package provided improved fidelity into the neutron energy distribution in the MeV range. In addition to the fluence monitor package irradiated with the samples, a dosimeter was also placed on the exterior of the assembly to estimate the perturbation in the fluence and accuracy of the MCNP modeled assembly. Figure 11 illustrates both the difference in the magnitude and energy distribution of neutrons inside and outside the assembly

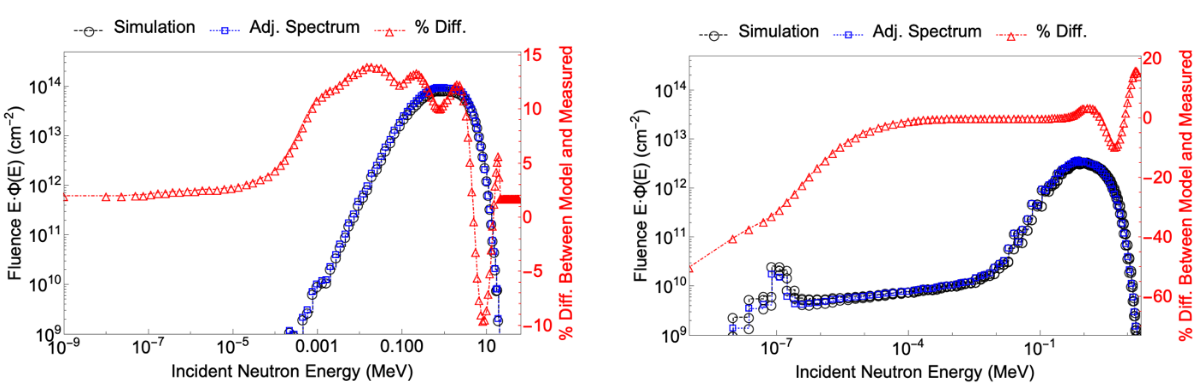

Figure 11: Neutron energy profile estimate inside and outside of the critical assembly as measured using a neutron dosimeter and modeled using MCNP.

In addition to the fluence characterization using neutron dosimetry and assortment of sensors are used to track the temporal dynamics of the pulse. A photo-multiplier tube and photo-diode are used to measure the power doubling period of the core and total length of the irradiation. As shown in Figure 12, the pulse dynamics between the ^{239}Pu and ^{237}Np irradiations did not differ significantly. The reactivity transient period was 14.5 and 14.7 µsec, respectively, and the irradiation lengths were statistically indifferent at around 54 µsec. These measurements assist in evaluating the energy of the measured fission yields and cross sections.

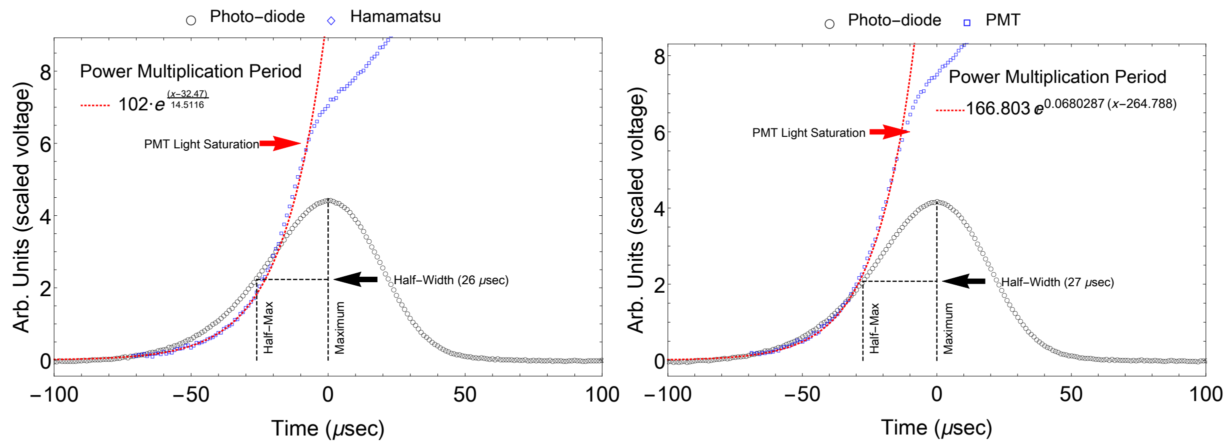

Figure 12: Pulse response observed during the ^{239}Pu (left) and ^{237}Np (right) experiments.

In the process of analyzing the ^{237}Np fission yield data, large contaminant gamma-ray peaks due to the decay of ^{238}Np were observed. Dr. Sean Burcher realized that the amount of ^{238}Np produced via the ^{237}Np(n,γ)^{238}Np reaction could be calculated by extrapolating the observed ^{238}Np activity back to the time of irradiation in Godiva. We deemed this was a worthwhile result to obtain and Dr. Burcher carried out the analysis. Through his efforts and in collaboration with Bruce Pierson from PNNL, who obtained the neutron flux spectrum, we have made a measurement of the integral cross-section of the ^{237}Np(n,γ)^{238}Np reaction with fission spectrum neutrons. The preliminary results of this cross-section measurement are shown in Figure 13, in comparison to previous measurements as well was the ENDF/B-VIII.0 and JEFF-3.3 evaluations. The final cross section results are under preparation for publication and will also be detailed in an internal technical report.

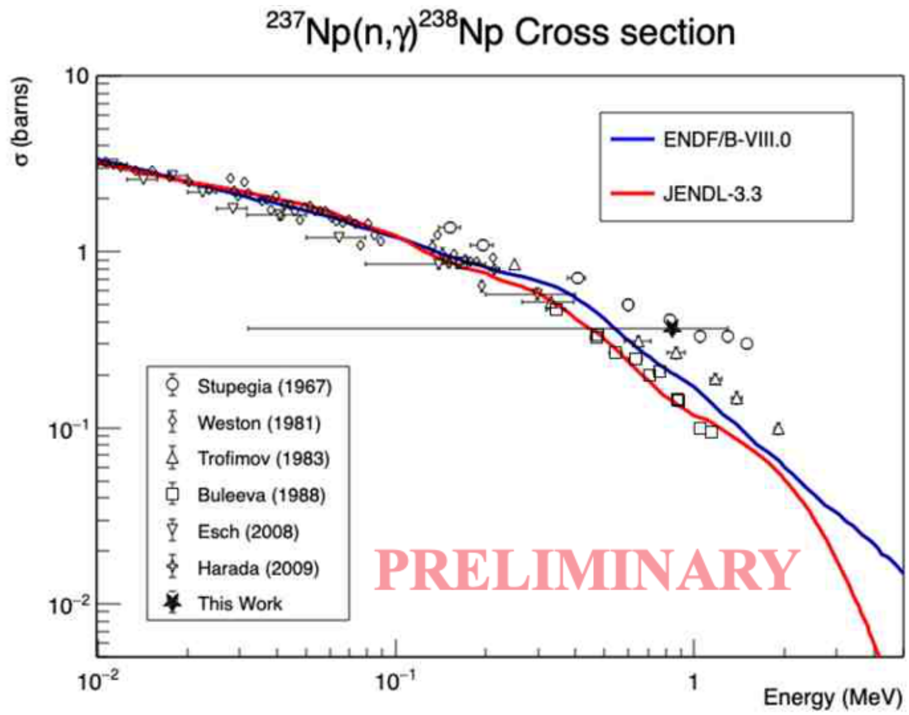

Figure 13: Preliminary result for the $^{237}Np(n,\gamma)^{238}Np$ cross section with fission spectrum neutrons.

The interim report has been completed and will be uploaded to WebPMIS imminently. The ^{237}Np report reference is, "Interim Report on Fission Product Yields from the Irradiation of ^{237}Np using the Godiva Critical Assembly", LLNL-TR-1023214, S. Burcher, et al. The dynamic behavior of the fission products can be seen in the cascade plot in Figure 14. Two things are apparent in this type of rendering of the data; 1) there can be gamma-ray peaks hidden by the Compton background if the measurement only provided a sum spectrum instead of list mode data which can be parsed into convenient sized time bins 2) without the resolution of high purity germanium a lot of detail would be lost to poor detector resolution of a NaI detector for instance. Both of these features of the measurement are key to being able to extract the maximum amount of information from a time dependent gamma-ray spectrum resulting from a prompt irradiation of an actinide target which induces fission. A very detailed accounting of all statistical and systematics uncertainties was also included in the report in keeping with our past standards.

Side note: Dr. Burcher had to remove the interfering gamma-rays from several of the ^{237}Np states in order to be sure we identified the correct low energy gamma-rays and did not assign them to the wrong nuclei. In doing so he realized that there were lines present from ^{238}Np, as mentioned previously. After some discussion with PNNL, we determined we could in fact obtain the neutron fluence accurately enough to then extract out the integral $^{237}Np(n,\gamma)^{238}Np$ cross section from the prompt neutron irradiation. A report and Physical Review C article are being worked on to document this result and outline what is required to obtain accurate integral (n,γ) cross sections from Godiva prompt burst-mode operations.

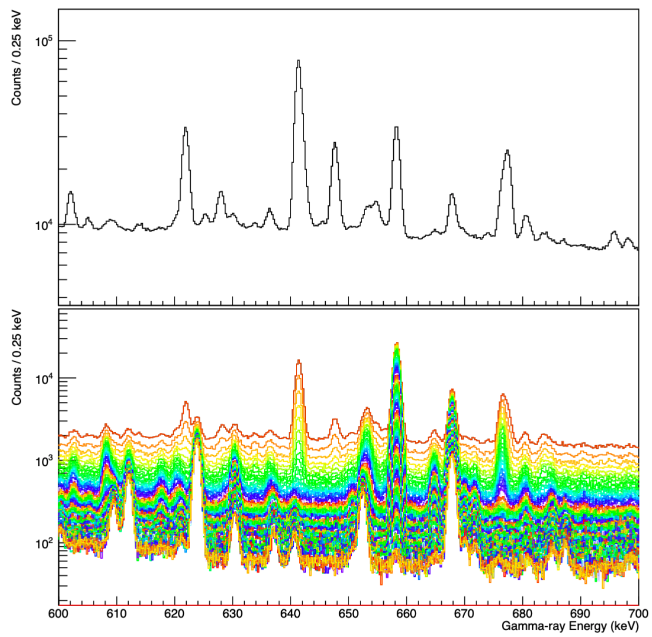

Figure 14: This is a cascade plot excerpted from the Burcher et al LLNL-TR-1023214 report on ^{237}Np. The top pane shows the sum spectrum one would observe by integrating for several hours. The bottom pane shows the same data displayed in 1-hour time bins. Since the data varies dramatically with time the cascade plot elucidates the temporal behavior of the fission product populations. It also allows one to see the growth and decay γ-ray peaks that would otherwise have been hidden in the sum plot (see 605 to 615 keV region).

Numerous fission product yields were obtained and are documented in the report; 20 short time scale (<4 hours observation point) isotopes were observed with 1 to 10 unique γ-rays, 25 long time scale (>4 hour observation point) with many confirmed with over half a dozen unique γ-ray lines. We may also have improved several half-lives and we also believe we have identified many cases where the underlying nuclear data γ-ray branching ratios are incorrect. This can be seen through the disagreement of only 1-2 γ-rays predicting a different FPY than the majority observed.

We will be checking for consistency in the branching ratio values by comparing all previous data using our updated analysis codes and looking for trends in the relative FPY predictions. We may also be able to extract out γ-γ coincidence data from the entire experiment and confirm the branching ratios.

Task 4: Precursor Fission Product Yields (LLNL)

Preparation for an experimental campaign at the Oregon State University (OSU) TRIGA reactor began in Q3. The experimental setup will consist of four Compton-suppressed High-Purity Germanium (HPGe) Clover detectors arranged in a cross-like formation. The Compton-suppression is achieved with bismuth germanate (BGO) shields. The γ-ray detection array will have a high efficiency to enable the detection of low activity fission products. In addition, the array will be highly segmented which will provide excellent γ-γ coincidence measurement capabilities. γ-γ coincidence data will be used to investigate branching ratios for γ-ray emission in short-lived fission products.

The DAQ electronics will consist of Mesytec shaping amplifiers coupled to CAEN analog-to-digital-converters (ADCs). The implementation of the DAQ still a work in progress and was stalled due to the COVID-19 shutdown. A full-scale test of the detection array and DAQ at LLNL, using various radioactive sources, is planned for early fall 2020.

In January 2020, Sean Burcher and Aaron Tamashiro traveled to OSU to begin the construction of the experimental setup and record background γ-ray spectra at the location of the setup to determine the feasibility of the measurement and assess the level of shielding required. A structure of lead bricks has been fully assembled and is shown in Figure 15. A single crystal HPGe detector was used to record a background γ-ray spectrum inside the lead shielding, and the background count rate was found to be ~250 Hz. From this measurement it has been determined that when scaled up to the full array efficiency and taking into account the veto from the BGO shields, the background rate will be well within acceptable range for the experimental campaign. The primary contributions to the background are the decay of ^{41}Ar and ^{16}N, both radioactive isotopes are produced via neutron capture reactions inside the reactor core and are present, to a small degree, throughout the reactor hall as a gas mixed into the ambient air.

The staff at the OSU TRIGA reactor are currently in the process of modifying the existing rabbit shuttle system to deliver samples from the reactor core to the new experimental setup location shown in Figure 15. The gap in the shielding at the top of the setup is to allow for the insertion of the pneumatic tube for the delivery of the rabbit shuttle. A custom low-profile, and cylindrically symmetric is currently being machined at OSU. This new terminus will be located in the center of the experimental setup, be able to stop the rabbit shuttle in a repeatable fashion, while also providing minimal attenuation of γ-rays from the irradiated sample.

The OSU TRIGA reactor restarted operations in June 2020. Once implementation of the DAQ system is completed, and a successful full-scale test of the detection array has been performed, the LLNL team will be ready to proceed with operations at OSU. The current timeline for the OSU experimental campaign is still uncertain due to the lab-wide travel restrictions, which are preventing travel to OSU.

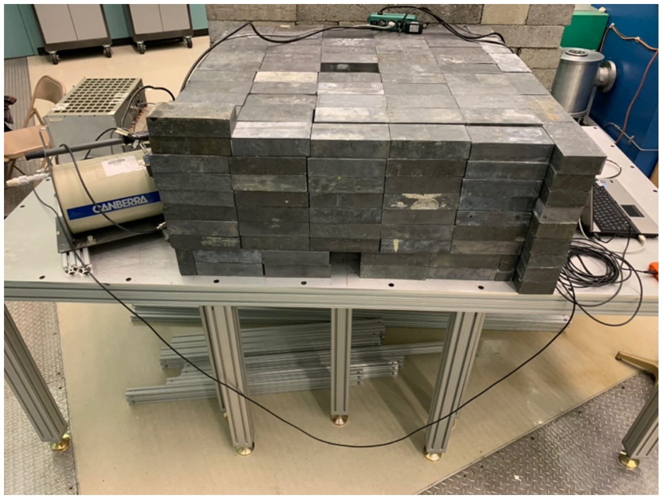

Figure 15: The beginning of the experimental setup at the OSU TRIGA reactor. The liquid nitrogen Dewar of a HPGe detector is visible protruding from the shielding on the left hand-side of the setup. The crystal was positioned at the approximate future location of the HPGe clover detectors in the full experimental setup. In this image the reactor containment vessel is approximately 20 feet to the right.

The new DAQ electronics were received in Q4, and are at LLNL. We have all the components needed to fully instrument the HPGe and Compton shield array. We have also been able to get approval to resume work on site with modified work schedules and some restrictions.

We have also formed a new national collaboration to share knowledge gained on how to use and program the new Mesytec electronics. The collaboration is called Project Enigma and involves 41 researchers from over 7 different institutions (US national labs and universities). We are working on setting up a website and software, manuals and lessons learned notes.

Task 5: Short-lived Assessment of Product Yields (LLNL)

In FY20 quarters 1 and 2, we began using the newly created database to search for fission products of interest. We will also evaluate some historical information and apply our new methods to it. More will be reported, as results are obtained, through a different channel. No work was performed in quarters 3 and 4 due to COVID related work restrictions.

Task 6: NCERC Experiment Support (LANL)

LANL personnel supported the execution of the ^{237}Np SLFPY irradiation on the Godiva assembly the week of January 20, 2020.

There was much activity planning for the CFPY run on Flattop that was originally rescheduled for the second week of September, 2020. Unfortunately, as the COVID restrictions continued into the summer this work shifted to rescheduling all NCERC experiments associated with this project in FY21. At this time, we have the following items on the official NCERC schedule.

1. Fission chamber detector tests on the Flattop-Oy assembly (Task #1) were scheduled for the week of November 16, 2020. Unfortunately, this had to be postponed due to a confluence of COVID related personnel shortages in several parts of LANL the prior two weeks. This prevented shipping the required equipment to NV in time.

2. Cumulative Fission Product Yield measurements on the Flattop-Oy assembly (Task #1) is scheduled for the week of April 12, 2021.
3. Activation Product measurements on the Flattop-Pu assembly (F2019 Task) is scheduled for the week of April 26, 2021.
4. The next Short-Lived Fission Product Yield measurement on the Godiva assembly (Task #3) has yet to be scheduled.

Task 7: NCERC Experiment Support (MSTS)

MSTS personnel supported the execution of the ^{237}Np SLFPY irradiation on the Godiva assembly the week of January 20, 2020.

MSTS has continued to support rescheduling of experiments in FY21.

FY21 Outlook

A major activity in early FY21 will be finalizing planning and execution of experiments that had to be rescheduled from FY20. This and many of the issues mentioned below are directly related to the very fluid nature of the COVID-19 pandemic.

General (All Tasks) Planned Activities for FY21

- Participation and presentation at the Workshop for Applied Nuclear Data Activities (WANDA 2021) meeting to be held as an extended WebEx from January 25 - February 3, 2021.
- Participation in the joint Schubert (Eiden) Review March 24-25, 2021.
- Continue engagement with theory and user communities.

Task 1: Cumulative Fission Product Yields (LANL/PNNL)

Planned Activities for FY21

Fission Chamber Development (LANL):

- Plan and execute detector/reference foil tests at the MIT Nuclear Reactor Lab.
- Plan and execute detector/reference foil tests at the NCERC.

Potential Project Impediments:

- Facility and resource scheduling at MIT and NCERC.
- Travel to the facilities and shipping and receiving of equipment and samples.

Sample Containment (LANL):

- Complete setup and testing of the Branson L20 with cold materials. Develop the procedure and safety basis for encapsulation of DU and HEU using the L20. Look into the safety requirements for using the L20 to encapsulate Pu metal macro-foils.
- Prepare DU and HEU macro-foils for the ^{235}U measurement at NCERC. The plan is to use the L20 for minimal encapsulation of these macro-foils.

Potential Project Impediments:

- Nothing specific.

Pu Sample Processing (LANL):

- Continue glovebox prep and method development for plutonium handling and processing. This is a requirement for the FY22 experimental plan.

Potential Project Impediments:

- Nothing specific.

Experiment Execution (LANL/PNNL):

- Complete analysis of data from the FY20 HEU measurement campaign (14 MeV). This will include generating all the measured R values along with the associated uncertainties for each measurement. Once compiled, the data will be shared between labs and discussed to identify similarities and differences.
- Planning and execution for the ^{235}U measurement campaign at NCERC (fission spectrum) to measure R-values and absolute fission product yields. This has been scheduled for the week of April 12, 2021.

Potential Project Impediments

- Facility and resource scheduling at LANL, PNNL and NCERC.
- Travel to the facility and shipping and receiving of equipment and samples.

Task 2: Cross Sections and Reaction Network Physics (LANL)

Planned Activities for FY21

- Complete assembly and calibration of the new vacuum chamber/SDD.
- Re-evaluate current experiment plan, and submit recommendations to HQ.
- Execute the approved experiment plan.

Potential Project Impediments

- Nothing specific.

Task 3: Short-lived Fission Product Yields (LLNL/PNNL)

Planned Activities for FY21

- Complete analysis of the ^{237}Np data.
- Prepare ^{233}U samples, schedule and execute the next measurement on the Godiva assembly. We are waiting on the final approvals to allow the commencement of wet chemistry operations in the specific rad-chem lab needed to fabricate the ^{233}U targets.

Potential Project Impediments

- Preparation of the ^{233}U samples.
- Facility and resource scheduling at LLNL, PNNL and NCERC.
- Travel to the facility and shipping and receiving of equipment and samples.

Project: Integral Measurements of Independent and Cumulative Fission Product Yields Supporting Nuclear Forensics and Other Applications

Task 4: Precursor Fission Product Yields (LLNL)

Planned Activities for FY21

- Complete setup and testing of the data acquisition system.
- Planning and execution for the measurement campaign at Oregon State University.

Potential Project Impediments

- Preparation of the ^{233}U samples.
- Facility and resource scheduling at LLNL, PNNL and NCERC.
- Travel to the facility and shipping and receiving of equipment and samples.

Task 5: Short-lived Assessment of Product Yields (LLNL)

Planned Activities for FY21

- Fold results from the ^{237}Np measurement into the ^{235}U, ^{238}U, ^{239}Pu FPY assessment.

Potential Project Impediments

- Nothing specific.

Task 6: NCERC Experiment Support (LANL)

Planned Activities for FY21

- Execute CFPY run on Flattop-Oy scheduled for the week of April 12, 2021.
- Execute API run on Flattop-Pu scheduled for the week of April 26, 2021.
- Schedule and execute SLFPY run on Godiva.
- Coordinate with task leads to plan and schedule FY22 activities.

Potential Project Impediments

- Facility and resource scheduling.

Task 7: NCERC Experiment Support (MSTS)

Planned Activities for FY21

- Support planned NCERC activities as scheduled.

Potential Project Impediments

- Facility and resource scheduling.

Meetings:

- Todd Bredeweg and Bruce Pierson attended US Nuclear Data Week 2018, held at Brookhaven National Laboratory the week of November 5. This annual meeting is the primary mechanism in the US to communicate and coordinate nuclear data activities and needs across academia, national labs and private industry. As such it is also an ideal venue to communicate and coordinate our activities with the broader theory and evaluation community.

- The Workshop for Applied Nuclear Data Activities (WANDA) was held at the Elliott School of International Affairs at the George Washington University from January 22-24, 2019, followed by a one-day invitation-only classified session on January 25, 2019. This workshop was modeled after the Nuclear Data Roadmapping Enhancement Workshop (NDREW), held at the UCDC center in 2018, that helped guide the 2018 Nuclear Data Interagency Working Group Funding Opportunity Announcement. The purpose of WANDA was to discuss nuclear data needs and potential solutions for nuclear energy, nonproliferation, isotope production, and stewardship science. The current project to study integral fission product yields was one of several fission product related efforts funded as part of the 2018 Interagency FOA, and it seemed appropriate to continue our participation in these efforts to coordinate investments in nuclear data research. Attendees from the Nuclear Data teams included Todd Bredeweg from LANL; Jason Burke from LLNL; and Larry Greenwood, Lori Metz and Bruce Pierson from PNNL. A final report from the workshop can be found at https://nucleardata.berkeley.edu/wanda/.

Publications

B.D. Pierson, A.M. Prinke, L.R. Greenwood, S.C. Stave, R.S. Wittman, J.G. Burch, J.T. Burke, S.W. Padgett, J.J. Ressler, G. Slavik, A. Tamashiro, A. Tonchev, W. Younes, “Improved Cumulative Fission Yield Measurements with Fission Spectrum Neutrons on 235U”, Nucl. Data Sheets 155, 86-97 (2019)